Specific heat and magnetization of $Co_{2-x}Sc_{1+x}Sn$

Z. W. Chen,[a] C. L. Lin, and T. Mihalisin
Department of Physics, Temple University, Philadelphia, Pennsylvania 19122

J. T. Wang
Department of Physics, Southern University and A&M College, Baton Rouge, Louisiana 70813

X. Q. Wang
Department of Materials Science and Engineering, University of Pennsylvania, Philadelphia, Pennsylvania 19104

Co_2ScSn crystallizes with the cubic $L2_1$ Heusler structure and is an itinerant ferromagnet with $T_c=238$ K. X-ray diffraction measurements show that $Co_{2-x}Sc_{1+x}Sn$ retains the $L2_1$ crystal structure with an essentially unchanged lattice constant for $0 \leq x \leq 0.15$. However, T_c determined from the magnetization measurements decreases monotonically with increasing Sc concentration from 238 K for $x=0$ to 40 K for $x=0.11$, and then remains at 40 K from $x=0.11$ to 0.14. The electronic specific heat coefficient γ is enhanced from 13 mJ/mole-K^2 for Co_2ScSn ($x=0$) to 30 mJ/mole-K^2 for $x=0.13$ ($Co_{1.87}Sc_{1.13}Sn$). In addition, the C/T versus T^2 plot shows a leveling-off behavior at low temperatures for the $x=0.13$ sample. The γ enhancement and level-off behavior observed when a system approaches a magnetic instability are discussed in terms of the self-consistent renormalization theory of spin fluctuations for weak itinerant ferromagnets and nearly magnetic systems. [S0021-8979(96)17208-3]

INTRODUCTION

Co_2ScSn crystallizes with the cubic $L2_1$ Heusler structure and is an itinerant ferromagnet with a saturation moment of 0.51 μ_B per Co atom.[1,2] We have shown[2] from magnetization measurements at low applied magnetic fields that the system undergoes a transition from a paramagnetic state to a helimagnetic state at $T_c=238$ K, and exhibits spin glass behavior below 100 K. The application of high magnetic fields suppresses the helimagnetic state and the spin glass behavior, and Co_2ScSn becomes a simple ferromagnet. We have also shown[3] that the substitution of Ni for Co in $(Co_{1-x}Ni_x)_2ScSn$ decreases T_c linearly with increasing Ni concentration and that T_c approaches zero in the vicinity of 40% Ni substitution ($x=0.4$). The electronic specific heat coefficient γ is enhanced from 13 mJ/mole-K^2 for Co_2ScSn ($x=0$) to 58 mJ/mole-K^2 for $x=0.4$ and $x=0.5$. In addition, for the $x=0.4$ and $x=0.5$ samples the specific heat divided by temperature (C/T) vs T^2 plot shows a leveling-off behavior at low temperatures, instead of the typical straight line behavior. γ enhancements and upturn behavior in C/T have been previously observed in f-electron systems, e.g. $(Ce,Gd)Sn_3$[4] and $(Y,U)Pd_3$,[5,6] when these systems are close to a magnetic instability. This behavior is also consistent with the predictions of self-consistent renormalization (SCR) theory[7] of spin fluctuations for nearly magnetic and weak itinerant magnetic systems.

Another way to drive the system towards a nonmagnetic state which does not involve the introduction of another element into the Co_2ScSn system is to depart from the 2 to 1 Co to Sc ratio, that is to make the $Co_{2-x}Sc_{1+x}Sn$ system. This system retains the $L2_1$ crystal structure for $x \leq 0.15$. The temperature dependence of the magnetization and specific heat and the T_c variation for this system are presented for $0 \leq x \leq 0.15$.

[a]Present address: Department of Materials Science and Engineering, University of Southern California.

EXPERIMENT

Polycrystalline $Co_{2-x}Sc_{1+x}Sn$ samples were prepared in an inert atmosphere arc furnace with appropriate care taken to compensate for the weight loss of the more volatile Sn. The samples were wrapped in Ta foils and were annealed under high vacuum at 800 °C for 8 weeks. This long period of annealing is necessary since we have shown[2,3] that the crystal structure and physical properties of Co_2ScSn and its related systems are significantly influenced by heat treatment. Powdered x-ray diffraction and high power, high-resolution electron microscopy measurements at room temperature were used to check sample homogeneity and crystal structure. The results indicated that $Co_{2-x}Sc_{1+x}Sn$ retains the cubic $L2_1$ Heusler structure with an essentially unchanged lattice constant of 6.190 Å for $0 \leq x \leq 0.15$. The magnetization was measured from 1.8 to 400 K at various magnetic fields up to 5.5 T using a commercial superconducting quantum interference device (SQUID) magnetometer. The specific heat was measured from 1.2 to 25 K in zero field and applied magnetic fields up to 4 T using a semiadiabatic heat pulse method.

RESULTS AND DISCUSSION

We have shown using magnetization, electrical resistivity, and ^{119}Sn Mossbauer measurements[2,8] that the ferromagnetic transition occurs at $T_c=238$ K for Co_2ScSn. Shown in Fig. 1 curve (a) is the temperature dependence of the magnetization of Co_2ScSn measured in an applied field of 20 Oe. It can be seen that the magnetization increases rapidly below 238 K. Below T_c, down to 100 K, the magnetization is essentially unchanged. This behavior of constant magnetization can be explained in terms of the helical magnetic state,[2] and

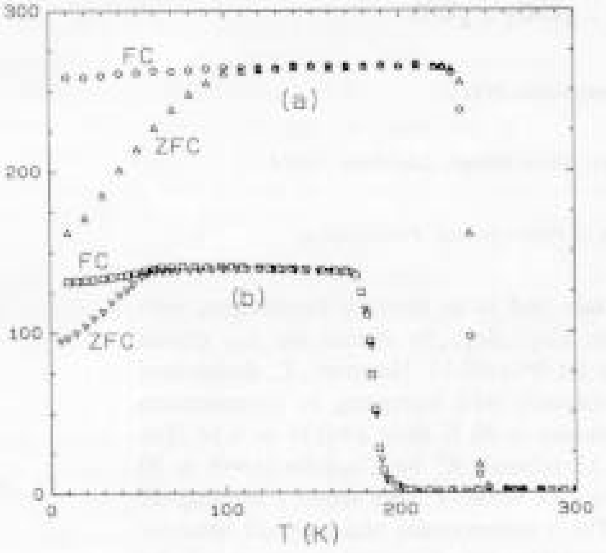

FIG. 1. The temperature dependence of the magnetization measured at $H=20$ Oe for (a) Co_2ScSn and (b) $Co_{1.97}Sc_{1.03}Sn$. ZFC: zero field cooled data, and FC: field cooled data.

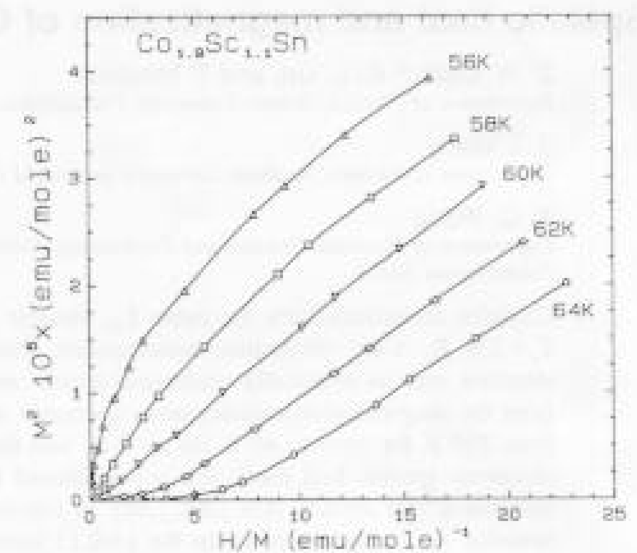

FIG. 2. Arrot plots (M^2 vs H/M) at $T=56$, 58, 60, 62, and 64 K for $Co_{1.9}Sc_{1.1}Sn$.

has also been observed for other itinerant magnets, e.g., MnSi and $Fe_{1-x}Co_xSi$.[9–11] Below 100 K the zero field cooled (ZFC) data increases monotonically with increasing temperature, but the field cooled (FC) data do not vary with temperature. This hysteretic behavior indicates that Co_2ScSn undergoes a second transition to a spin glass like state at low temperatures, i.e., below 100 K. We have also shown[2] that the helical magnetic state and spin glasslike behavior are suppressed by the application of high magnetic fields, and Co_2ScSn becomes a simple ferromagnet.

We have shown[3] that T_c decreases linearly with decreasing Co concentration in the $(Co_{1-x}Ni_x)_2ScSn$ system, and approaches zero in the vicinity of 40% Ni substitution for Co ($x=0.4$). We have also measured the temperature and magnetic field dependence of the magnetization for the $Co_{2-x}Sc_{1+x}Sn$ system with $0 \leq x \leq 0.15$. The magnetization measured at low applied fields for the samples with lower Co concentrations, i.e., $0<x\leq 0.15$, shows a similar behavior to that shown in Fig. 1 curve (a). Curve (b) in Fig. 1 shows the FC and ZFC magnetization for $Co_{1.97}Sc_{1.03}Sn$ ($x=0.03$), also measured at $H=20$ Oe. It can be seen, however, that T_c and the spin glass transition for this sample occur at lower temperatures, i.e., 190 and 55 K, respectively. Again, the helical magnetic state and spin glass behavior for these samples with lower Co concentrations can be also suppressed by the application of high magnetic fields (not shown), and the samples then become simple ferromagnets.

In order to extract the ferromagnetic transition temperature T_c, we have measured M versus H for several temperatures near T_c for every sample reported here. Arrott plots (M^2 versus H/M) for $Co_{1.9}Sc_{1.1}Sn$ ($x=0.1$) at various temperatures are shown in Fig. 2. T_c is determined as the temperature for which a linearly extrapolated curve on the Arrott plot goes through the origin. We find a T_c value of 60 K from this plot for this sample. T_c determined from Arrott plots is in good agreement with T_c defined as the temperature where $|dM/dT|$ is maximum.

Shown in Fig. 3 is T_c versus Sc concentration $(1+x)$ for the $Co_{2-x}Sc_{1+x}Sn$ system. T_c decreases monotonically with decreasing Co concentration (increasing Sc content $1+x$) from $T_c=238$ K for Co_2ScSn ($x=0$) to 40 K for $x=0.11$. It is interesting that T_c remains almost the same (40 K) for the samples with even higher Sc concentrations in the region between $(1+x)=1.11$ and 1.14. One might argue that Co content can not be lowered below 1.89 (or that the true maximum value of $(1+x)$ is 1.11), and that is why T_c stays unchanged for $0.11<x\leq 0.15$. For the following reasons we rule out this speculation. First, for the samples with $0.11<x\leq 0.15$ no detectable extra phases are seen via the x-ray diffraction measurements. Second, we did not find a contribution to the magnetization from free Co metal or magnetic impurity phases. Third, we observe that the value of the mag-

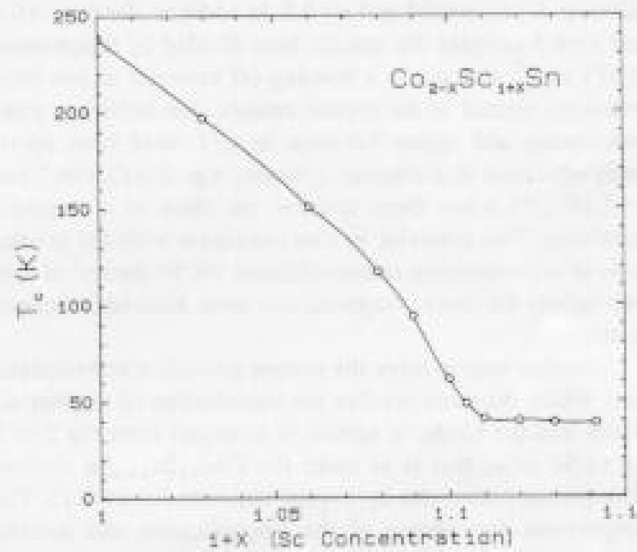

FIG. 3. T_c as a function of Sc concentration $(1+x)$ for the $Co_{2-x}Sc_{1+x}Sn$ system with $0\leq x\leq 0.15$.

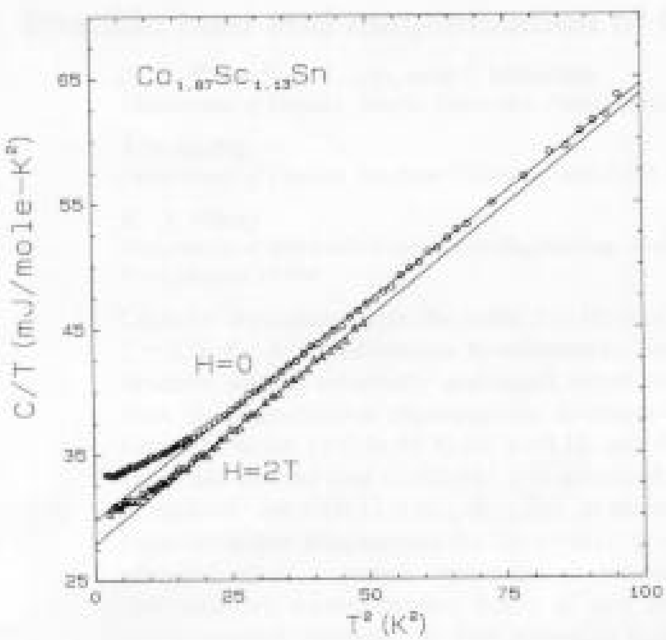

FIG. 4. C/T vs T^2 plot measured at applied fields $H=0$ and $H=2$ T for $Co_{1.87}Sc_{1.13}Sn$. The straight line indicates a linear fit to higher temperature data from $T=5$ to 10 K.

netization at low temperatures decreases monotonically as one lowers the Co content across the entire regime, including $0.11<x\leq 0.15$.

As mentioned earlier, the variation of γ as the system approaches a magnetic instability is of primary interest. Shown in Fig. 4 is the C/T versus T^2 data measured at $H=0$ and 2 T for $Co_{1.87}Sc_{1.13}Sn$. For $H=0$ γ values of 33.5 and 30 mJ/mole-K^2 are obtained from C/T at $T=1.4$ K and from a linear fit of higher temperature data ($T=5$–10 K), respectively. Either of the γ values mentioned above for $Co_{1.87}Sc_{1.13}Sn$ is larger than that of Co_2ScSn, i.e., $\gamma=13$ mJ/mole-K^2. This enhanced γ behavior when the system approaches a magnetic instability has been reported previously for many f- and d-electron systems. For example, γ is greatly enhanced for 10% Gd, 20% U, and 60% Co in the $(Ce_{1-x}Gd_x)Sn_3$,[4] $(Y_{1-x}U_x)Pd_3$,[5,6] and $(Co_{1-x}Ni_x)_2ScSn$[3] systems, respectively.

It is clear that the C/T versus T^2 plot for $Co_{1.87}Sc_{1.13}Sn$ shown in Fig. 4 shows a leveling-off behavior at lower temperatures. This is behavior has also been observed in the cases of heavy fermion f-electron systems,[4–6] enhanced paramagnetic itinerant d-electron systems[12] and weakly magnetic d-electron systems.[13] We do not believe that the leveling-off behavior seen in C/T versus T^2 for $Co_{1.87}Sc_{1.13}Sn$ is due to the presence of a spin glass transition for two reasons. First a plot of T_g (spin glass transition temperature) versus $1+x$ (not shown here) shows that T_g goes to zero for $1+x\sim 1.07$. Second this type of C/T versus T^2 behavior is not seen for samples which have a spin glass transition namely pure Co_2ScSn and $Co_{2-x}Sc_{1+x}Sn$ with $1+x<1.07$. The self-consistent renormalization (SCR) theory of spin fluctuations[7] can explain the magnetic, electric, and thermodynamic properties of nearly magnetic and weakly magnetic itinerant systems. According to the prediction of the SCR theory, an additional contribution of the form $T[\ln(T/T_{sf})]$ should be added to the specific heat resulting in a leveling-off behavior in a C/T versus T^2 plot at low temperatures. Although the applied field of 2 T reduces the specific heat over the entire temperature range, it is not sufficiently strong to suppress the spin fluctuations, and thus the leveling-off behavior in $H=2$ T remains similar to that in zero field. That is, one expects that T_{sf} is considerably larger than 2 K which would correspond to the field of 2 T.

In conclusion, we have measured the magnetization and specific heat of the $Co_{2-x}Sc_{1+x}Sn$ system with $0\leq x\leq 0.15$. T_c decreases monotonically with decreasing Co concentration from 238 K for $x=0$ to 40 K for $x=0.11$ and then remains at 40 K between $x=0.11$ and $x=0.14$. The specific heat data for $Co_{1.87}Sc_{1.13}Sn$ show that γ is enhanced to 30 mJ/mole-K^2 and the C/T versus T^2 plot exhibits a leveling-off behavior at low temperatures.

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[1] S. K. Malik, P. L. Paulose, R. Nagarajan, A. E. Dwight, P. P. Vaishnava, and C. W. Kimball, Hyperfine Inter. **34**, 427 (1987).
[2] C. L. Lin, T. Mihalisin, and N. Bykovetz, J. Magn. Magn. Mater. **116**, 355 (1992).
[3] C. J. Fuller, Z. W. Chen, N. Anbalagan, C. L. Lin, and T. Mihalisin, J. Appl. Phys. **73**, 5427 (1993).
[4] C. L. Lin, T. Yuen, G. Y. Jiang, and T. Mihalisin, J. Appl. Phys. **70**, 6092 (1991).
[5] C. L. Seaman, M. B. Maple, R. W. Lee, S. Ghamaty, M. S. Torikachvili, J. S. Kang, L. Z. Liu, J. W. Allen, and D. L. Cox, Phys. Rev. Lett. **67**, 2882 (1991).
[6] B. Andraka and A. M. Tsvelik, Phys. Rev. Lett. **67**, 2886 (1991).
[7] T. Moriya, in *Spin Fluctuations in Itinerant Electron Magnetism*, Springer Series in Solid State Sciences 56, (Springer, Berlin, 1985).
[8] Z. W. Chen, C. L. Lin, T. Mihalisin, and N. Bykovetz, J. Appl. Phys. **73**, 6974 (1993).
[9] K. Shimizu, H. Maruyama, H. Yamazaki, and H. Watanabe, J. Phys. Soc. Jpn. **59**, 305 (1990).
[10] J. Beille, J. Voiron, F. Towfiq, M. Roth, and Z. Y. Zhang, J. Phys F **11**, 2153 (1981).
[11] J. Beille, J. Voiron, and M. Roth, Solid State Commun. **47**, 399 (1983).
[12] M. Shiga, K. Fujisawa, and H. Wada, J. Phys. Soc. Jpn. **62**, 1329 (1993).
[13] C. J. Fuller, C. L. Lin, T. Mihalisin, F. Chu, and N. Bykovetz, Solid State Commun. **83**, 863 (1992).